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## EUROPEAN PATENT APPLICATION

(21) Application number : **94870067.9**

(51) Int. Cl.<sup>5</sup> : **C08F 279/02, C08L 51/04,  
// (C08L51/04, 51:04)**

(22) Date of filing : **15.04.94**

(30) Priority : **15.04.93 US 48632  
30.04.93 US 56323**

(43) Date of publication of application :  
**19.10.94 Bulletin 94/42**

(84) Designated Contracting States :  
**BE DE ES FR GB IT NL SE**

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(54) **High impact polystyrene having improved gloss and impact characteristics.**

(57) A composition and process for making it are disclosed which teach multimodal particle size high impact monovinylaromatic polymer material having good gloss properties as well as high impact strength. The bimodal particle size rubber component of the material utilizes a small amount of a very large particle size rubber and a large amount of a very small particle size rubber. The unique and advantageous method of manufacturing the material involves introducing pre-existing pellets of a rubber/polymer blend into a monomer solution also containing a rubber capable of producing particles of a different range of sizes than that of the rubber component in the pellets, and polymerizing the monomer/dissolved pellet mixture into a multimodal particle size high impact material.

EP 0 620 236 A1

FIELD OF THE INVENTION

The present invention relates to thermoplastic compositions utilizing polymers of monovinylaromatic compounds which have been modified with rubber to increase their impact strength. More particularly the present invention discloses a high impact polystyrene material which is particularly advantageous for use in manufacturing items that normally must be manufactured from an ABS (acrylonitrile butadiene styrene) type of material.

The present invention discloses a high impact polystyrene material which utilizes a bimodal particle size distribution (BPSD) rubber component which is added to the monovinylaromatic compound by unique and novel processes to obtain a new and highly useful product exhibiting increased gloss and impact resistance.

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BACKGROUND OF THE INVENTION

It is well-known that rubber reinforced polymers of monovinylaromatic compounds, such as styrene, aliphatic methyl styrene, and ring-substituted styrenes are desirable for a variety of uses. More particularly, rubber-reinforced polymers of styrene having included therein discrete particles of a cross-linked rubber, for example, polybutadiene, the discrete particles of rubber being dispersed throughout the styrene polymer matrix, can be used in a variety of applications including refrigerator linings, furniture, household appliances, and toys. The conventional term for such rubber-reinforced polymers is "High Impact Polystyrene" or "HIPS".

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The physical characteristics and mechanical properties of HIPS are dependent upon many factors, including the particle size of the cross-linked rubber particles. One of the most important characteristics of HIPS materials is the ability of such materials to exhibit a high surface gloss which is aesthetically appealing while also maintaining a high impact strength in order to be desirable for use in articles such as furniture and appliances.

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It has been known that properties of HIPS such as impact and gloss are strongly affected by rubber particle size and distribution at a given rubber concentration. Unfortunately, these two main properties counteract each other and the result is that the larger rubber particle utilized to obtain strength and impact detracts from the appearance and gloss of the final material. It has been known since the late 1970's that a small number of large rubber particles when combined with a large number of small rubber particles and blended into a HIPS material synergistically achieve a HIPS with high impact and high gloss. The phrase commonly associated with utilizing a combination of large and small rubber particles is "Bimodal Particle Size Distribution" (BPSD) HIPS.

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BPSD HIPS is normally prepared by one of three known methods. The first such method is melt-blending, the second is "parallel polymerization", and the third is "split feed polymerization".

In melt-blending, two HIPS materials of different particle size distributions are blended together in an extruder or a mixer.

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In parallel polymerization, two independently polymerized mixtures, one of which has small rubber particles and the other of which has large rubber particles are combined prior to final polymerization in a third polymerization reactor after phase inversion (particle formation), and then the combined mixture undergoes final polymerization.

"Split feed polymerization" involves the process wherein approximately 70 to 95% of the feed solution is polymerized past the point of phase inversion, and then the remainder of the feed solution is added to the reactor preventing phase reinversion. The rubber particles in the second feed are immediately formed upon addition and dispersed, resulting in much larger particles than the initial rubber particles. The large rubber particles have a unique morphology wherein a large number of tiny occlusions and a relatively thick rubber membrane are formed.

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One of the earliest patents recognizing the advantage of bimodal particle size distribution rubber HIPS was that granted to Lanza, U.S. Patent No. 4,153,645 dated 1979 which discloses an improved balance of properties obtained from melt blends comprising 65 to 85% HIPS, having small rubber particles (approximately 0.6  $\mu\text{m}$ ) and about 15 to 35% HIPS having large rubber particles (2-5  $\mu\text{m}$ ) which were prepared by means of mass-suspension polymerization. Lanza stressed that the selection of the proper ratio of HIPS materials, followed by dilution with crystal polystyrene to obtain rubber level control, would provide an optimum balance of properties. The addition of 20% HIPS material having 5  $\mu\text{m}$  rubber particles to a HIPS material having 0.6  $\mu\text{m}$  rubber particles increased Izod impact strength from 1.8 up to 4.6 ft. lb./in. (96 to 246 J/m) when the total rubber concentration was 10.5%.

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In 1980, the Lavengood patent, U.S. Patent No. 4,214,056 disclosed a method for preparing melt-blends exhibiting improved toughness and gloss, with a 70 to 95% HIPS material having 0.5 to 1  $\mu\text{m}$  rubber particles and the remaining HIPS material having 2 to 3  $\mu\text{m}$  rubber particles. The two HIPS materials were prepared in a multistage stirred reactor. When a 10% HIPS material of 2.5  $\mu\text{m}$  rubber particle size was blended with 90% HIPS having 0.6  $\mu\text{m}$  rubber particles, with a total rubber concentration of 7 to 8%, the Izod increased from 0.9 up to 1.8 ft.lb./in. (48 to 96 J/m) without degradation of gloss, which was measured at 96 according to ASTM

523 test method (60° angle).

Japanese Patent, JP 63,112,646 discloses a material having a good balance of strength, stiffness, toughness and uniform color obtained from utilizing a 75% HIPS material having rubber particles exhibiting a core-shell morphology and a particle size of 0.1 to 0.6  $\mu\text{m}$ , combined with 25% HIPS having 0.7 to 2.0  $\mu\text{m}$  rubber particles with a "salami" morphology, and 0.05% polydimethylsiloxane. This HIPS material achieved an Izod strength of 2.2 ft.lb./in. (117 J/m) compared to 0.7 ft.lb./in. (37 J/m) without the bimodal particle size distribution. The resulting material as in the previous two patented processes utilized melt-blending to achieve the final product.

Examples of "parallel polymerization" BPSD HIPS include the material taught by the Dupre patent, U.S. Patent No. 4,146,589, dated 1979.

In this patent, BPSD HIPS materials were manufactured in a continuous process by means of a CSTR (continuous stirred tank reactor) utilizing parallel polymerization.

A HIPS comprising 70 to 95% of 0.5 to 1.0  $\mu\text{m}$  rubber particles and 5 to 30% of 2 to 3  $\mu\text{m}$  rubber particles was manufactured by parallel polymerization and resultantly increased the Izod approximately 50% while maintaining a relatively high gloss. In one example, Izod and gloss of one material, which was 90% 0.7  $\mu\text{m}$  rubber particles, and 10% 2.2  $\mu\text{m}$  rubber particles were respectively 1.8 ft.lb./in. (96 J/m) and 95 with a total rubber concentration of 8 to 9%. A similar invention was disclosed by the Aerts, et al. patent, EP 0 412 801 A1, published in 1991, where BPSD HIPS was prepared in plug-flow reactors using linear flow stirred tower reactors and parallel polymerization. The patent claimed that this method achieved advantages in controlling rubber particle size, the degree of grafting, and the molecular weight of the resulting product compared to HIPS materials prepared by various other methods. HIPS material comprising 50 to 95% of 0.2 to 0.7  $\mu\text{m}$  rubber particles and 5 to 50% of 1.5 to 5  $\mu\text{m}$  rubber particles exhibited improved mechanical and gloss properties utilizing a rubber for the small particles, which was a block copolymer of styrene and butadiene (40/60). In one example, a HIPS material comprising 80% of 0.2  $\mu\text{m}$  rubber particles and 20% of 3.7  $\mu\text{m}$  rubber particles had an Izod value of 3.0 ft-lb/in (160 J/m) and a gloss of 91.

Another example of "parallel polymerization" HIPS material manufacture is found in the Echte patent, U.S. Patent No. 4,493,922, issued in 1985. This patent discloses HIPS compositions having 60 to 95% of 0.2 to 0.6  $\mu\text{m}$  small particles and 5 to 40% of 2 to 8  $\mu\text{m}$  large particles and claimed a balance of gloss and impact properties. The morphologies of the small and large particles were different and were respectively, capsule morphology and cell/coil morphology. The patent stated that this BPSD HIPS could be prepared also by blending (melt/solution) or by parallel polymerization.

Examples of "split feed polymerization" can be found in patents such as that issued to Mott, et al. in 1980 (U.S. Patent No. 4,221,883) which disclosed a method for producing a BPSD HIPS by means of using split feed polymerization, where the second feed was added to the polymerizing first feed when the percent solids was greater than 30. The patent claimed that polymers prepared by this method, when compared to others, were generally more uniform in tensile strength and further exhibited better flow characteristics during molding operations. The patent also claimed materials which possessed a good combination of gloss and tensile and elongation.

The Dupre patent, U.S. Patent No. 4,334,039 issued in 1982 disclosed a process for preparing BPSD HIPS which allegedly showed an unexpected increase in toughness without degradation of gloss or flow properties. The Dupre process disclosed a first feed solution polymerized up to 10 to 40% conversion and a second feed solution added to the reactor simultaneously with the pre-polymerized first feed solution. The resulting products contained 70 to 95% of 0.5 to 1.0  $\mu\text{m}$  rubber particles and 5 to 30% of 2 to 5  $\mu\text{m}$  rubber particles. The patent also claimed that small rubber particles formed in the process have a longer residence time which reportedly increased the percent grafting and toughness.

The Damen, et al. European patent, EP 0 096 447 A2, published in 1983, purported to disclose a BPSD HIPS which was manufactured by adding the second feed solution which was either a fresh solution or a partially polymerized but non-inverted solution, to the pre-polymerized first feed, wherein small particles were dispersed. The patent alleged that the process resulted in a BPSD HIPS having 70 to 95% rubber particles of 0.5 to 1.5  $\mu\text{m}$  and 5 to 30% rubber particles of 4 to 10  $\mu\text{m}$ , where the percent graft of small particles was at least 130 and the resulting material exhibited a balance of properties such as gloss, elongation and impact strength. The patent specifically claimed a gloss of at least 80, an elongation of at least 30, and an impact strength in J/m of at least 14 times the percent butadiene in the rubber.

The Japanese patent, JP 336617, dated 1989, purportedly describes a BPSD HIPS material having 65 to 95% of 0.1 to 0.5  $\mu\text{m}$  rubber particles and 5 to 35% of 0.5 to 2.0  $\mu\text{m}$  rubber particles.

The Burk patent, U.S. Patent No. 4,254,236, claims to disclose a modified "split-feed polymerization" process wherein large rubber particles were prepared in a second feed solution by dissolving polystyrene to induce phase inversion. The second feed solution containing preformed large rubber particles was fed into the pre-

5 polymerized first feed solution when the first feed solution was at 15 to 50% conversion level. It was alleged in the Burk patent that the size of the large particles could be more easily controlled because the size of the second rubber particles could be increased by increasing the concentration of the dissolved polystyrene in the second feed solution. HIPS material prepared by this method containing 50 to 95% of 0.5 to 1.5  $\mu\text{m}$  rubber particles and 5 to 50% of 2 to 10  $\mu\text{m}$  rubber particles allegedly provided a balance of impact strength and gloss properties.

### SUMMARY OF THE INVENTION

10 The present invention provides a process that is unique and novel which may be utilized to manufacture a bimodal rubber particle size distribution HIPS material which more easily controls the parameters of the material such as gloss and impact strength and does so much more efficiently than those known processes described in the aforementioned prior art. The process and material of the present invention are achieved by the utilization of a unique method of introducing rubber particles into the polystyrene material. This process is categorized as "pellet-feed solution polymerization". In pellet-feed solution polymerization, a small amount of HIPS having a large particle component is dissolved in styrene along with a rubber, which will give smaller rubber particles than those of the dissolved HIPS. Then the solution is polymerized via a normal polymerization process. The principle of this polymerization is that the cross-linked rubber particles in the pellet-feed do not lose their size by reinversion, but maintain their size throughout their flow through the reactors. As a result, the HIPS material formed will have a bimodal particle size distribution. This rubber reinforced polystyrene material demonstrates a high gloss and high impact strength that can be more closely controlled than the complicated systems of the three aforementioned blending processes.

15 20 Also, the economics of the present system are considerably more beneficial than those of the prior three systems and the resulting material exhibits properties equal to or better than those materials disclosed in the aforementioned prior art.

25 In addition to the unique and novel pellet-feed process, the present invention also discloses BPSD HIPS materials manufactured according to conventional processes such as the melt blend process, which exhibits superior gloss and impact strength, and which utilize a smaller percentage of large particle size rubber particles than those materials disclosed in the prior art and wherein the particle size of the large component is larger than the particle size of the conventional materials. This results in improved properties over the disclosed materials made according to the previous processes.

### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

30 35 The present invention discloses a thermoplastic composition containing a polymer of a monovinylaromatic compound which has been modified with a bimodal rubber particle size distribution to increase the material's impact strength and surface gloss. The compound may be obtained by utilizing conventional methods, such as melt blending or alternatively by the process of "pellet-feed polymerization", wherein the finished HIPS pellets having large rubber particle size are dissolved in a rubber solution which will yield a small particle size HIPS material resulting in a bimodal particle size distribution in the finished product. The small amount of large rubber particles, having a size of 5 to 10  $\mu\text{m}$  and a content of less than 5% added to a HIPS material of approximately 40 0.5  $\mu\text{m}$  rubber particles yields a synergistic effect on the gloss and impact strength. The most efficient and economical method of manufacturing such material is the aforementioned pellet-feed solution polymerization.

### DESCRIPTION OF THE FIRST PREFERRED EMBODIMENT

45 In a first preferred embodiment, the BPSD HIPS material of the present invention was manufactured utilizing conventional manufacturing techniques such as melt blending.

46 In this preferred embodiment, a high gloss, high impact material was manufactured by melt blending a HIPS material utilizing a small rubber particle component having a particle size of 0.3 to 0.5  $\mu\text{m}$  and a core-shell morphology with a HIPS material having a large rubber particle component with particle sizes in the range of 50 8 to 9  $\mu\text{m}$ . The large particle size rubber component comprised 2 to 4% of the entire rubber content of the material and the small rubber particle component comprised the remainder of the rubber content in the final material.

55 The final material was actually manufactured by utilizing two separate HIPS materials, each having the desired particle size contained therein. One HIPS material had the small rubber particle size and the second HIPS material had the large rubber particle size. The two HIPS materials were melt blended in a single screw extruder having a temperature profile as follows : Zone 1-163°C (325°F)/Zone 2-177°C (350°F)/Zone 3-204°C (400°F)/Zone 4-204°C (400°F)/extruder die-218°C (425°F). A Malvern particle size analyzer was used to determine the particle size distribution, i.e., the volume average particle size (mean, medium) and the percent under X micron size. The Izod impact was determined using compression molded or injection molded samples,

depending upon the quantity of the samples. The samples were prepared according to ASTM D256 test method. The gloss was measured on injection molded samples utilizing 60° angle according to ASTM 523 test method.

Tables I and II hereinbelow indicate the results of the melt blending of two different rubber particle size HIPS materials to obtain a BPSD HIPS material as the final result.

In the following examples, indicated by the results shown in Tables I-III, the following test methods were utilized: IZOD ASTM D-256 (values in J/m calculated from values in ftlb/in); Gloss ASTM D-523-85 at 60°; and Gardner Impact ASTM D-3029 and D-1709, using a Gardner "Coverall" Mandrel Impact Tester.

Table I illustrates the blending of a small particle size HIPS material having an average rubber particle size of 0.4 µm with two large rubber particle size HIPS materials, one with a rubber particle size of 5.5 µm and the other with a rubber particle size of 8 µm.

TABLE I

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	A (0.4 µm)	B (5.5 µm)	C (8 µm)	RPS (Rubber Particle Size) Volume median (µm)	Izod (ftlb/in)	Gloss 60°
20	100	0	0	0.3	1.2	64
	97	3	0	0.4	1.5	80
	95	5	0	0.5	2.1	112
	0	100	0	5.5	2.0	107
25	98	0	2	0.3	1.8	96
	95	0	5	0.5	2.3	123

A = Small particle size HIPS material

B = 5.5 µm HIPS material

C = 8 µm HIPS material

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TABLE II

	Volume Median				Izod	Gloss <sup>a</sup>
	D (0.5 µm)	E (8 µm)	F (9 µm)	RPS (µm)	(ftlb/in)	(J/m)
40	100	0	0	0.5	0.8	43
	98	2	0	0.6	1.7	91
	97	3	0	1.6	2.3	123
45	96	4	0	3.6	2.9	155
	95	5	0	4.4	3.1	165
	97	0	3	2.1	2.2	117

<sup>a</sup>2-3 units lower than previous measurements.

50 D = Pilot Plant HIPS using 12% rubber\* in polystyrene.

E = HIPS material C (8 µm)

F = HIPS material C (9 µm)

55 It can be seen from examples C and F in Tables I and II above, that very good Izod impact and Gloss values can be achieved using high percentage (95-97) of small rubber particle HIPS (0.4-0.5 µm) melt-blended with a small percentage (3-5) of a very large rubber particle HIPS (8-9 µm).

DESCRIPTION OF THE SECOND PREFERRED EMBODIMENT

Table III below represents test runs of BPSD HIPS manufactured according to a heretofore unused method of making such products which method is referred to herein as the "pellet feeding process" or "pellet solution polymerization". Rather than using one of the conventional bimodal particle size HIPS manufacturing processes, such as melt-blending, "parallel polymerization", or "split feed polymerization", the present embodiment introduces a unique process whereby existing pellets of HIPS material having a single range of particle sizes is introduced into a styrene feed solution which contains a rubber material which will yield a different particle size range than that of the HIPS pellets. The HIPS pellets used, preferably having the large particle size producing rubber components, are dissolved in styrene monomer along with a small particle size producing rubber. This feed solution is then polymerized via commercial polymerization methods.

The principle involved in this unique process is believed to be based upon the apparent fact that the cross-linked rubber particles in the HIPS pellets are not reinverted when the pellets are dissolved in the styrene monomer prior to polymerization. Therefore, these rubber particles maintain their original size and may even increase in size to some extent, perhaps as a result of additional grafting and cross-linking, during the polymerization process. This, when combined with the smaller rubber particles obtained from the rubber dissolved in the monomer feed, achieves a desirable bimodal distribution.

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TABLE III

		Izod (ftlb/in)	Gardner	Gloss 60°
25	HIPS A' + 2% HIPS B			
	1. melt blending	0.9	48	4
	2. pellet feed	1.2	64	29
30	HIPS A' + 5% HIPS B			
	1. melt blending	1.2	64	63
	2. pellet feed	1.3	69	70
35	HIPS B' + 2% HIPS C			
	1. melt blending	1.6	85	95
	2. pellet feed	1.7	91	118
40	HIPS B' + 3% HIPS C			
	1. melt blending	2.3	123	106
	2. pellet feed	2.0	107	121
45	HIPS C' + 2% HIPS D' + 3% HIPS F'			
	1. dry blend	1.2	64	87
	2. pellet feed	1.7	91	151
50	HIPS E' + 4% HIPS C			
	1. dry blend	2.4	128	112
	2. pellet feed	2.8	149	140
	HIPS A': Contains 10.5% rubber (0.5 µm)			
	HIPS B': Contains 12% rubber (0.5 µm)			
	HIPS C': Contains 14% rubber (0.4 µm)			
	HIPS D': Contains (2µm)			
	HIPS E': Contains 14% rubber			
	HIPS F': 4 µm HIPS material			

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In Table III above, various combinations of HIPS components were utilized to manufacture BPSD HIPS

samples using first a conventional blending process (sample #1), and then using the same components in a "pellet feed solution polymerization" process (sample #2). From this table it is evident that the Pellet Feeding Process generally provides improved Izod impact, Gardner impact, and gloss (60°) over conventional HIPS blending techniques such as melt-blending. In every sample except the fourth, every property was equal to or better in the pellet feeding process than in the melt-blending process. In the fourth sample, Gardner impact and gloss was improved in the pellet feeding sample but Izod impact was reduced.

Thus the "pellet feed solution polymerization" process of manufacturing BPSD HIPS materials is preferable to conventional processes, providing improved qualities in the finished HIPS material. In addition, this embodiment of the invention provides further advantages:

- 10 (1) No additional capital investment is required to shift from single particle size HIPS to bimodal particle size HIPS, whereas the conventional bimodal processes all require costly additional equipment to manufacture BPSD HIPS;
- 15 (2) By being able to closely control the amount of pellets added into the polymerization system, the manufacturer has the ability to maintain very exact control of the rubber particle size distribution, since the content of particles in the pellet portion of the feed stays constant; and,
- 20 (3) Transition materials or selected off-grade HIPS materials may be used to supply the large particle component, thereby further reducing costs.

Thus the advantages of the two embodiments of the present invention can be clearly demonstrated. A new composition of BPSD HIPS has been disclosed herein which utilizes a small percentage of rubber particles with very large sizes, and a high percentage of small particle size rubber particles to obtain a high-gloss, high impact HIPS material.

Also, a unique and easily controlled process of making a BPSD HIPS material is taught which utilizes existing HIPS pellets, which are added with a rubber to styrene monomer prior to or during polymerization resulting in superior high impact polystyrene.

25 Although a specific preferred embodiment of the present invention has been described in the detailed description above, the description is not intended to limit the invention to the particular forms of embodiments disclosed therein since they are to be recognized as illustrative rather than restrictive and it will be obvious to those skilled in the art that the invention is not so limited.

For example, whereas the first embodiment is disclosed utilizing melt-blending, one skilled in the art could 30 adapt the invention to utilize other processes such as "parallel polymerization." Also, whereas the second embodiment utilizes preformed HIPS pellets, other forms of HIPS material such as granules, fines, rods, films, and others could be utilized in conjunction with or as replacements for the pelletized HIPS. Additionally, whereas one of the rubber particle sizes is formed from a rubber dissolved in the styrene monomer, it is clear that the monomer-feed solution could be rubber-free and all the particle size rubber components could be added 35 by utilizing several existing rubber-modified polystyrene materials of different particle sizes, with the result being multi-modal particle size high-impact polystyrene.

Thus, the invention is declared to cover all changes and modifications of the specific example of the invention herein disclosed for purposes of illustration which do not constitute departure from the spirit and scope of the invention.

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## Claims

1. A process for manufacturing a high-gloss, high impact polystyrene material having a multi-modal rubber 45 particle size distribution, said method comprising :
  - preparing a polymerization feed solution of styrene monomer and a rubber in solution, said rubber yielding a predetermined first particle size;
  - selecting a high impact polystyrene material having a rubber component with a second particle size different from said first rubber particle size;
  - dissolving said high impact polystyrene material in said feed solution; and,
  - polymerizing said feed solution into a bimodal particle size distribution high impact polystyrene.
2. The process of Claim 1, wherein said first rubber particle size is between 0.1 and 1.0  $\mu\text{m}$ .
3. The process of Claim 1, wherein said second rubber particle size is between 3 and 15  $\mu\text{m}$ .
4. The process of Claim 1, wherein said first rubber particle size is 0.1 to 1.0  $\mu\text{m}$  and said second rubber particle size is 3 to 15  $\mu\text{m}$ .

5. The process of Claim 1, wherein said polystyrene material is a pelletized high impact polystyrene and said polymerization feed solution comprises rubber dissolved in styrene monomer.

6. The process of Claim 3, wherein said polystyrene material is a pelletized high impact polystyrene and said feed solution is rubber dissolved in styrene monomer.

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7. A method of controlling the rubber particle size distributions of a multi-modal high impact polystyrene material, said method comprising :

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- preparing a feed solution comprising a first particle size yielding rubber dissolved in styrene monomer;
- dissolving in said feed solution a first supply of high impact polystyrene material containing a second rubber particle size;
- dissolving in said feed solution at least one additional supply of high impact polystyrene material having at least one additional size of rubber particle, different from said first and second rubber particle sizes; and,
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- polymerizing said feed solution and dissolved polystyrene materials into multi-modal rubber particle size high impact polystyrene.

8. The method of Claim 7, wherein said first particle size yielding rubber has a particle size of 0.1 to 1.0  $\mu\text{m}$  and said at least one additional size of rubber particle is 3 to 15  $\mu\text{m}$ .

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9. A method of controlling the rubber particle size distributions of a multi-modal high impact polystyrene material during the manufacture thereof, said method comprising :

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- dissolving in a styrene monomer feed solution a single particle size yielding rubber material;
- dissolving in said feed solution at least one high impact polystyrene material containing rubber particles of different size than said single particle size yielding rubber; and,
- polymerizing said feed solution into multi-modal high impact polystyrene.

10. The method of Claim 9, wherein said single particle size yielding rubber is of a small particle size, and said at least one high impact polystyrene material comprises a polystyrene having a large rubber particle.

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11. The method of Claim 9, wherein said single particle size yielding rubber is of a small particle size, and said at least one high impact polystyrene material comprises a first polystyrene having a large rubber particle and a second polystyrene having a medium size rubber particle between the small particle size and the large particle size.

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12. A bimodal rubber particle size high impact monovinylaromatic polymer material exhibiting high gloss and impact strength, said polymer material comprising :

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- a first monovinylaromatic polymer containing a small rubber particle component having a particle size of 0.3 to 0.5  $\mu\text{m}$  and a core-shell morphology; and
- a high impact second monovinylaromatic polymer blended with said small rubber particle polymer, said monovinyl-aromatic material having a large rubber particle component with particle sizes in the range of 8 to 9  $\mu\text{m}$ ;
- wherein said large rubber particle component comprises 2 to 4 percent of the entire rubber content of the finished product.

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13. The monovinylaromatic polymer of Claim 12, wherein said first and second monovinylaromatic polymers are each a high impact polystyrene.

14. The monovinylaromatic polymer of Claim 13, wherein said first and second polymers are melt-blended together in an extruder.

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15. A high gloss, high impact blended polystyrene composition comprising a polystyrene matrix and a bimodal rubber particle size component in said matrix, said rubber component having a first particle size distribution of large particles in the range of 8 to 9  $\mu\text{m}$  in diameter, and a second particle size distribution of small particles in the range of 0.3 to 0.5  $\mu\text{m}$  in size, with the large particles comprising 2 to 4 percent of the total rubber component, and the remainder being the small particle size rubber component.

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European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number

EP 94 87 0067

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.CLS)
A	US-A-4 277 574 (M.B. JASTRZEBSKY ET AL.) * column 9, line 35 - line 68; claims 1-3,19 *	1,5,7,15	C08F279/02 C08L51/04 //(C08L51/04, 51:04)
A	EP-A-0 460 541 (BASF) * page 4; claim 1; example 1 *	1,12-15	
A,D	DE-A-26 20 579 (LABOFINA) * page 4, paragraph 2; claims 1,3,5; table 1 *	1,12-15	
A	WO-A-85 02192 (DOW) * page 15, line 20 - line 30; claim 1; examples; table II *	1,15	
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TECHNICAL FIELDS SEARCHED (Int.Cl.5)			
C08F C08L			
The present search report has been drawn up for all claims			
Place of search		Date of completion of the search	Examiner
THE HAGUE		1 August 1994	Engel, S
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	

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